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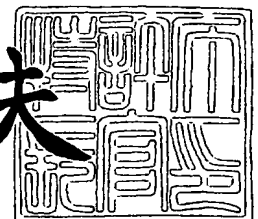
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【ブルーフの要否】 要

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1. TITLE OF INVENTION

INJECTION MOLDED PRODUCT MADE OF BIODEGRADABLE
AROMATIC POLYESTER BLEND COMPOSITIONS AND METHOD OF
MANUFACTURE THEREOF

2. Claims

1. An injection molded product made of a molding composition comprising (A) 20 to 98.8wt. at least one polyester being selected from the group consisting of :

an aromatic polyester copolymer (a) having repeating units comprising an acid component and a glycol component, wherein the acid component comprises about 50 to 90 mol% terephthalic acid, about 0.2 to about 6 mol% sulfonic acid metal salt, and about 4 to 49.8 mol% aliphatic dicarboxylic acid; wherein the glycol component comprises about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol% diethylene glycol;

a polyester copolymer (b) prepared by copolymerization with said copolymer (a) with polyalkylene glycol,

a branched polyester copolymer (c) prepared by polycondensation of said copolymer (a) with polyalkylene glycol, and,

a polyester copolymer (d) having repeating units comprising aromatic dicarboxylic acids and glycol component; with the proviso that the mol% of said aromatic dicarboxylic acids of said polyester copolymer is less than the mol% of the carboxylic acid content of said copolymers (a), (b), and (c),

(B) 1 to 60 wt% material selected from the group consisting of reinforcing and filling materials,

(C) 0.1 to 7 wt% crystallization promoter, and,

(D) 0.1 to 5 wt% lubricant.

2. The injection molded product of claim 1 in which the molding composition further comprises 1 to 60 wt% inorganic flame retardant.

3. The injection molded product of any one of claim 1 or claim 2 in which the molding composition is a blend of said copolymers (a), (b), (c) and (d).

4. The injection molded product of any one of claims 1 to 3 wherein said product has heat distortion at temperature not lower than 80°C and the crystallization speed is faster than 1.2min. at 120°C.

5. The injection molded product of any one of claims 1 to 3 wherein the molded product is biodegradable.

6. A method for manufacturing an injection molded product comprising the steps of:

(I) blending

(A) 20 to 98.8 wt% at least one polyester being selected from the group consisting of :

an aromatic polyester copolymer (a) having repeating units comprising an acid component and a glycol component, wherein the acid component comprises about 50 to 90 mol% terephthalic acid, about 0.2 to about 6 mol% sulfonic acid metal salt, and about 4 to 49.8 mol% aliphatic dicarboxylic acid; wherein the glycol component comprises about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol% diethylene glycol;

a polyester copolymer (b) prepared by copolymerization with said copolymer (a) with polyalkylene glycol,

a branched polyester copolymer (c) prepared by polycondensation of said copolymer (a) with polyalkylene glycol, and,

a polyester copolymer (d) having repeating units comprising aromatic dicarboxylic acids and glycol component; with the proviso that the mol% of said aromatic dicarboxylic acids of said polyester copolymer is less than the mol% of the carboxylic acid content of said copolymers (a), (b), and (c);

(B) 1 to 60 wt% material selected from the group consisting of reinforcing and filling materials,

(C) 0.1 to 7 wt% crystallization promoter, and,

(D) 0.1 to 5 wt% lubricant

(II) injection molding said molding composition prepared by said blending.

7. The method of claim 6 in which the molding composition further comprises 1 to 60 wt% at least one flame retardant selected from inorganic flame retardant, phosphorous-based flame retardant and phenolic polymer..

3. Detailed Description of the Invention

The present invention relates to an injection molded product formed of a molding composition comprising at least one polyester selected from the group consisting of a first aromatic polyester copolymer consisting repeating units derived from terephthalic acid, a metal salt of sulfonic acid, aliphatic dicarboxylic acid, ethylene glycol and diethylene glycol, copolymers prepared by copolymerization or polycondensation of the first polyester with polyethylene glycol and the polyester comprising relatively less aromatic dicarboxylic acids than that of the other polyesters to be blended and glycol, which is biodegradable and has extremely desirable mechanical strength, heat resistance and moldability resulting into being highly suitable for many various applications subject to disposal, if desired, flame resistance and also relates to a method of manufacture thereof.

DESCRIPTION OF RELATED ART

Prompted by societal concerns and demands concerning the disposal of plastic products such as plastic films, research is being done on biodegradable resin compositions. Active efforts are underway to develop biodegradable aromatic polyester resin compositions which are degradable under the high-humidity, high-temperature conditions associated with waste composting processes. Various uses are being proposed for such compositions. For example, Tokuhyo Hei 5-507109, 6-505513 and 6-505040 describe polyesters prepared by polymerizing a glycol component made of ethylene glycol and diethylene glycol with two acid components; namely, an alkali metal or alkaline earth metal salt of a sulfonic acid, and terephthalic acid. These prior-art references also describe fibers, films, sheets and fiber nonwoven fabrics composed of such polyesters.

Tokukai 2001-172487 describe biodegradable moldings with inorganic fillers that are vacuum molded from aliphatic polyester sheet. Such moldings have improved modulus, thermal resistance and impact resistance.

Tokukai Hei 9-169897 describe biodegradable sheet with natural fibers. Such sheet has enough strength and heat resistance during use.

Such thermoplastic materials including polyolefin, polyester, polyamide and polyvinyl chloride are useful in molding articles by any of the techniques commonly

used with thermoplastic materials, e.g., compression molding, injection molding, extrusion, blow molding. However, in certain applications such as daily necessities, electronics appliance, industrial applications and automobile applications, products made from biodegradable polymer would be desirable to have mechanical strength and heat resistance not less than those of products made from such other thermoplastic materials. It is also well known that addition of fillers into aliphatic polyester molding compositions may improve heat resistance and modulus of elasticity, but may not improve molding cycle, mold release and moldability enough to meet with requirements from the above applications. As noted above, aliphatic polyester resins have improved strength and heat resistance by adding fillers. Yet, they continue to lack sufficient stiffness, impact resistance and heat resistance for practical use such as daily necessities, electronics appliance, industrial applications and automobile applications. Moreover, when aliphatic polyester with fillers are injection molded, it causes low moldability such as low mold releasability or long molding cycle.

To overcome these shortcomings, an injection molded article is formed from an aromatic polyester copolymer consisting repeating units derived from terephthalic acid, a metal salt of sulfonic acid, aliphatic dicarboxylic acid, ethylene glycol and diethylene glycol, enhancing performance properties, in particular heat resistance and moldability includes reinforcing and filling materials, crystallization promotor and lubricant to facilitate crystallization of the aromatic polyester molding composition resulting into improvement of moldability. .

BRIEF SUMMARY OF THE INVENTION

It is therefor an object of the invention to provide an injection molded product made of a molding composition comprising:

(A) 20 to 98.8 wt% at least one polyester being selected from the group consisting of :

an aromatic polyester copolymer (a) having repeating units comprising an acid component and a glycol component, wherein the acid component comprises about 50 to 90 mol% terephthalic acid, about 0.2 to about 6 mol% sulfonic acid metal salt, and about 4 to 49.8 mol% aliphatic

dicarboxylic acid; wherein the glycol component comprises about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol% diethylene glycol;

a polyester copolymer (b) prepared by copolymerization with said copolymer (a) with polyalkylene glycol,

a branched polyester copolymer (c) prepared by polycondensation of said copolymer (a) with polyalkylene glycol, and,

a polyester copolymer (d) having repeating units comprising aromatic dicarboxylic acids and glycol component; with the proviso that the mol% of said aromatic dicarboxylic acids of said polyester copolymer is less than the mol% of the carboxylic acid content of said copolymers (a), (b), and (c),

(B) 1 to 60 wt% material selected from the group consisting of reinforcing and filling materials,

(C) 0.1 to 7 wt% crystallization promoter, and,

(D) 0.1 to 5 wt% lubricant.

Other embodiments of the invention include inorganic flame retardant. The invention also contemplates injection molded products formed of a blend of said polymers (a), (b), (c) and (d). In a preferred embodiment of the injection molded product of the invention has a heat distortion at temperature not lower than 80° C, and crystallization speed of the molding composition is faster than 1.6min at 120deg.C. The invention further provides injection molded product that is biodegradable.

According to a further aspect of the invention, there is provided a method for manufacturing an injection molded product comprising the steps of:

(I) blending

(A) 20 to 98.8wt% at least one polyester being selected from the group consisting of:

an aromatic polyester copolymer (a) having repeating units comprising an acid component and a glycol component, wherein the acid component comprises about 50 to 90 mol% terephthalic acid, about 0.2 to about 6 mol% sulfonic acid metal salt, and about 4 to 49.8 mol% aliphatic dicarboxylic acid; wherein the glycol component comprises about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol% diethylene glycol;

- a polyester copolymer (b) prepared by copolymerization with said copolymer (a) with polyalkylene glycol,
- a branched polyester copolymer (c) prepared by polycondensation of said copolymer (a) with polyalkylene glycol, and,
- a polyester copolymer (d) having repeating units comprising aromatic dicarboxylic acids and glycol component; with the proviso that the mol% of said aromatic dicarboxylic acids of said polyester copolymer is less than the mol% of the carboxylic acid content of said copolymers (a), (b), and (c);
- (B) 1 to 60 wt% material selected from the group consisting of reinforcing and filling materials,
- (C) 0.1 to 7 wt% crystallization promoter, and,
- (D) 0.1 to 5 wt% lubricant
- (II) injection molding said molding composition prepared by said blending.

DETAILED DESCRIPTION OF THE INVENTION

The injection molded product of the invention, the molding composition and method of manufacturing thereof are described in detail hereinafter.

The injection molded product of the invention is formed of the molding composition comprising at least one aromatic polyester copolymer selected from the group consisting of the polyester copolymers given below, reinforcing or filling material, a crystallization agent and lubricant, optionally other ingredients. All described below.

Aromatic polyester copolymers

An aromatic polyester copolymer (a) has repeating units comprising an acid component and a glycol component. The acid component is about 50 to 90 mol %, and preferably, about 52 to 83 mol %, terephthalic acid. The copolymer has repeating units comprising an acid component and a glycol component. The acid component is about 50 to 90 mol %, and preferably, about 52 to 83 mol %, terephthalic acid.

Further according to the present invention, the acid component is about 0.2 to 6 mol%, and preferably about 2 to 5 mol%, sulfonic acid metal salt. Illustrative examples of the sulfonic acid metal salt include metal salts of 5-sulfoisophthalic acid, metal salts of 4-sulfoisophthalic acid, and metal salts of 4-sulfophthalic acid. Of these, metal salts of 5-sulfoisophthalic acid are preferred. Preferred examples of the metal

ions include ions of alkali metals such as sodium, potassium and lithium, or of alkaline earth metals such as magnesium. The most preferred sulfonic acid metal salt is the sodium salt of 5-sulfoisophthalic acid.

The sulfonic acid metal salt is not only relatively expensive, when used in excess it renders the polyester water-soluble and moreover affects physical characteristics.. The sulfonic acid metal salt significantly contributes to the degradability of the molded products using the composition of this invention even at a low content of 0.2 mol %. Further according to the present invention, the acid component is about 4 to 49.8 mol %, and preferably about 10 to 45 mol %, aliphatic dicarboxylic acid. At less than 4 mol %, the glass transition temperature cannot be significantly lowered. On the other hand, an aliphatic dicarboxylic acid level in excess of 49.8 mol % invites a decline in the glass transition temperature, causing a loss of suitable stiffness in the molded products

The aliphatic dicarboxylic acid preferably has 2 to 18 carbons, and more preferably 2 to 10 carbons. Illustrative examples include azelaic acid, succinic acid, adipic acid, sebacic acid and glutaric acid. Of these, glutaric acid is preferred.

Composting that involves the degradation of molded articles is typically carried out under high-temperature, high-humidity conditions. Because this is generally done at a temperature of about 70°C or less. The aromatic polyester molding compositions of the present invention have a glass transition temperature (T_g) not higher than preferably about 70°C, and especially about 65°C. In the invention, an aliphatic dicarboxylic acid is used to set the glass transition temperature not higher than about 70°C. An ester-forming derivative of the dicarboxylic acid, such as the dimethyl ester, may be used in place of the dicarboxylic acid.

The glycol component is about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol % diethylene glycol, and preferably about 80 to 98 mol % ethylene glycol and about 2 to 20 mol % diethylene glycol. More than 50 mol % of diethylene glycol units adversely influences the mechanical properties of the film, such as the tensile strength, whereas less than 0.1 mol % results in poor degradability.

The glass transition temperature may be further lowered by substituting up to 20 mol % of the ethylene glycol with another glycol such as triethylene glycol. A balanced range of properties, especially mechanical properties and biodegradability, can be achieved by preparing the injection molded products of the present invention

by choosing selected amounts of respective components within the above mol% ranges. If it is desired to enhance the mechanical properties of the molded products, the amount of terephthalic acid is increased, and if it is desired to enhance biodegradability, the amount of aliphatic dicarboxylic acid is increased, resulting in a lower glass transition temperature (T_g).

A polyester copolymer (b) is prepared by copolymerization with the copolymer (a) with polyethylene glycol. Polyethylene glycol component is about 0.1 to 20wt.%.

A branched polyester copolymer (c) is prepared by polycondensation of the copolymer (a) with polyethylene glycol. Minor amounts of polyfunctional branching agents, such as trimellitic acid, are incorporated to branch polyethylene glycol to modify melt rheology and film processing. The amount of polyethylene glycol component is about 0.1 to 20wt.%.

Polyethylene glycol used for the copolymers (b) and (c) may be replaced with other polyalkyleneglycol such as polypropylene glycol. Mw should be relatively low to achieve biodegradation and processability.

A polyester copolymer comprises acid components comprising aromatic dicarboxylic acids with less amount than that of the aromatic carboxylic acids contained in any of the copolymers (a), (b), and (c), and glycol component. For example, Polybutylene succinate, polylactide, polycaprolactone could be applied.

The weight proportion of the aromatic dicarboxylic acids in the composition of the fourth polyester of the invention should be from 0-70 parts by weight per hundred parts of the polyester, preferably 0-50 parts by weight.

The molding composition of the present invention will comprise 20 to 98.8wt%, preferably 40 to 90 wt%, and most preferably, 50 to 80 wt% of the polyester copolymers based on the total weight of the polyester copolymers, reinforcing or filling materials, crystallization promoter and lubricant. (To Mizutani san, can you fill in blank ?)

Reinforcing or filling materials

The composition of the present information can also contain any reinforcing or filling material universally known to the world. For instance, cited may be glass fiber, carbon fiber, potassium titanate, whisker, kaolin, talc, graphite or aramid fibers, glass

beads, aluminum silicate, wollastonite, asbestos, calcium carbonate, barium sulfate, mica and the like, and combinations of such materials.

The composition of the present information can also contain natural reinforcing or filling materials such as starch, hemp, flax, cotton, pulp, cellulose, diatom, wood powder, rayon and the like, and combinations of such materials. Natural reinforcing materials can also be wastes such as wood, paper, cacao, tea leaves, soy bean, bamboo, dried garbage and the like.

The compounding ratio of reinforcing or filling materials can be selected arbitrarily in accordance with the application of moldings such as desired mechanical characteristics and the molding shape. Generally, it is in the range of 1~60wt, preferably 20 ~ 50 wt %, most preferably 20-40wt.%. When the amount of fillers compounded is less than 1 wt %, sufficient improvement in mechanical characteristics cannot be obtained. On the other hand, when the amount of fillers compounded exceeds 60 wt %, the flow property of the melted composition obtained falls causing poor moldability.

A crystallization promoter

The crystallization promoter should be such that it contains at least one of alkaline metal ion source, alkaline-earth metal ion source, zinc ion source, aliphatic amide and talc in sufficient quantity. Preferably for alkaline, alkaline-earth, zinc ion source, the crystallization promoter is an organic hydrocarbon acids containing between about 7 and 54 carbon atoms or organic polymers having at least one carboxylic group.

The amount of crystallization promoter is in the range of 0.1-7.0wt.%, preferably 0.5 to 5 wt.%. When the amount of crystallization promoter is less than 0.1 wt.%, sufficient crystallization could not be obtained during the molding process and affects moldability. When the amount of crystallization promoters are more than 7 wt.%, it will affect the mechanical characteristics.

Lubricant

The composition of the present information will contain any well-known lubricant that can contribute to crystallization of aromatic polyester copolymers in the present invention. For instance, aliphatic ester such as pentaerythrityl tetrastearate,

stearyl stearate, dipentaerythritol and pentaerythrityl distearate, olefin wax, paraffin wax, natural wax and silicone wax can be applied. Pentaerythrityl tetrastearate is preferred. Generally the amount of lubricant is 0.1 to 5 wt.%, preferably 0.5 to 2 wt.%.

Flame Retardant

A common method of imparting flame resistance to thermoplastic polyester resin compositions involves adding a halogenated organic compound as a flame retardant along with an antimony compound that acts as a synergist for the flame retardant. However, the use of halogenated flame retardants has certain drawbacks in that these materials exhaust trace amount of toxic gases on combustion and tend to corrode the barrels of compounding extruders, the surfaces of molding machines, and other equipment they come in contact with at elevated temperatures. Thus, flame retardants which has less effect on environment and machine was addressed in this patent.

The composition of the present information may contain non halogenated flame retardants. Inorganic flame retardant, Phosphorous-based flame retardant, phenolic polymer, thermo plastic acrylic type polymer and silicone type flame retardant can be used. Inorganic flame retardants include, but not limited to, $Mg(OH)_2$, $Al(OH)_3$, $CaCO_3$ and $BaSO_4$. Inorganic flame retardant is most preferred due to the environmental friendliness.

The phosphorus-based flame retardant may be organic or inorganic. Suitable inorganic flame retardants include, but are not limited to, red phosphorus and aluminum and zinc phosphonate salts. Suitable organic phosphorus-based flame retardants include phosphonates, phosphates, and oligomeric and polymeric phosphates. A preferred flame retardant is resorcinol bis(di-2,6-xylyl)phosphate, which is described in Japanese Kokai H9-143350, and is a low cost product marketed under the name PX-200 by Daihachi Chemicals Co., Japan. The phosphorus-based flame retardant should be present in about 0 to about 25 weight percent based on the total weight of the composition. The phenolic polymer may include novolacs or resols. These may be partially or fully cured by heating and/or the use of cross-linking agents. Preferred are novolacs. More preferred are novolacs that do not have added cross-linking agents and are not heat reactive. There is no particular limitation as to the

form to be used: pulverized, granular, flake, powder, acicular, liquid, and other forms are suitable. The phenolic polymer may be used as a blend of two or more types. Phenolic polymer synthesized from renewal resources such as wood can also be used. In the present invention, the amount of phenolic polymer used should be about 0 to about 25 weight percent based on the total weight of the composition.

Polymerization process

The aromatic polyester polymer used to form the inventive moldings can generally be prepared by any well-known polymerization method. For example, a straight-chain polyester in which the monomer units are randomly distributed along the molecular chain can be prepared by charging a polymerizer with all of the above monomer constituents together with antimony or some other catalyst, and carrying out polycondensation under suitable polycondensation conditions. Another method that may be used involves initially reacting two or more of the monomer constituents to prepare a prepolymer, then adding the remaining monomer constituents and polymerizing.

The aromatic polyester polymer used to form the moldings of the invention decomposes under the high-humidity, high-temperature conditions typical of composting. Most of the monomer and oligomer (i.e., terephthalic acid, glycol, and oligomers thereof) which forms as a result of such degradation is readily digested by microorganisms in the solid wastes or compost, ultimately becoming carbon dioxide and water.

Other additives

Conventional additives such as plasticizers, , toughening agents, , nucleating agents, anti-electrification agents, flame retardant, antioxidants, heat stabilizer, dye and pigment, UV stabilizer and weathering stabilizers may be added to the foregoing aromatic polyester polymer for the purpose of adjusting the moldability or mechanical properties, provided the mechanical characteristics, degradability and other properties critical to the polyester are not altered thereby, and the resulting aromatic polyester composition subjected to moldings formation.

The polyester copolymer that forms the inventive moldings typically has an intrinsic viscosity within a range of 0.1 to 1.5, and preferably 0.3 to 1.2.

Blending process

The resin composition used in the present invention can be obtained usually by melt-blending the copolyester, reinforcing or filling materials, crystallization promotor, lubricants and the above-mentioned additives optionally with a usual melt-mixer such as monoaxial or biaxial extruder, Banbury mixer, kneader or mixing roll. The entire or part of the components to be compounded may be supplied to the melt-mixer simultaneously or separately. The most general method is to dry-blend components in advance followed by melt-kneading with the above-mentioned melt-mixer to homogenize and forming pellets. The pellet-shaped resin composition thus prepared is usually kept in the sufficiently dried state and charged into the molding machine hopper for molding.

The aromatic polyester moldings of the invention is well-suited for use in a range of applications, including agricultural and horticultural supplies such as plant pot for farming and gardening use, any kinds of daily necessities such as the handle of toothbrush, containers, dishes, cutleries, and even to automobile parts or office automation equipment parts.

Injection Molding

The method further comprises the step of forming injection moldings above blend of aromatic polyester polymer. The process of forming moldings involves feeding blend of aromatic polyester polymer-containing flakes to an extruder, melting the flakes, and extruding the melt through a nozzle to the mold die. The molten plastic is cooled, crystallized and solidified in the mold, which is kept closed. By heating the mold to proper temperature at about 120 °C, the plastic is crystallized and achieves high heat resistance. Then the mold is opened to eject the solid plastic molded article.

The described compositions characterized by the unexpected faster speed of crystallinity and improvement of heat resistance and moldability can also be applied to other molding process including crystallization step therein, such as sheet forming, vacuum molding, injection blown bottle and direct blown bottle.

EXAMPLES

Examples are given below by way of illustration, although the examples are not intended to limit the scope of the invention.

TEST METHODS

The methods of measurement and evaluation used in the examples are described below.

Moldability

Good: Sufficient crystallization and mold release

Poor: poor moldability due to insufficient crystallization and mold release.

Tensile strength:

Measured in accordance with ASTM D638

Elongation:

Measured in accordance with ASTM D638

Flexural strength:

Measured in accordance with ASTM D970

Flexural Modulus:

Measured in accordance with ASTM D970

Izod Impact strength:

Measured in accordance with ASTM D256.

Heat distortion temperature:

Measured in accordance with ASTM D648

Compost Degradability:

Test pieces of a given size were placed in compost for 15 weeks, following which the specimens were visually examined.

YES: Shape readily breaks down under outside forces

NO: No change

t_{max}

Crystallization speed were evaluated by DSC with rapid cooling unit which is capable of achieving the maximum cooling rate of 200°C/min.. Isothermal measurement was carried out to observe crystallization peak. Each isothermal curve at measurement temperature (120°C) was obtained after rapid cooling from 220°C. For the samples evaluated, the time of maximum crystallization t_{max} was collected to show the isothermal properties.

Flame resistance:

UL Test No. UL-94 (20 mm Vertical Burning Test) using 1/8th inch (referred to in the Table 2 as 3.2 mm) thick test pieces.

EXAMPLES

In the examples hereafter, copolyester 1 means an aromatic polyester copolymer(density, 1.35 g/cm³; melting point, 200°C; melt index at 220°C under 2,160 g of loading, 11 g/10 min) having repeating units composed of an acid component that is about 50 to 90 mol % terephthalic acid, about 0.2 to 6 mol % sodium 5-sulfoisophthalate and about 4 to 49.8 mol % glutaric acid, and a glycol component that is about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol % diethylene glycol.

Copolyester 2 means an aromatic polyester copolymer(density, 1.35 g/cm³; melting point, 200°C; melt index at 220°C under 2,160 g of loading, 28 g/10 min) prepared by copolymerization with the copolyester 1 with 1 to 20 wt.% polyethylene glycol.

Copolyester 3 means an branched aromatic polyester copolymer(density, 1.35 g/cm³; melting point, 185°C; melt index at 220°C under 2,160 g of loading, 23 g/10 min) copolymerizing copolymer1 with 1 to 20wt.% polyethylene glycol.

In Table 1 and 2 ,each material name stands for,

GF : glass fiber, St.Na: sodium stearateMon.Na:Sodium MontanatePTS:pentaerythrityl tetrastearate ,

PX-200: resorcinol bis(di-2,6-xylyl),

Phenolic polymer : Novolac HRJ12700CP manufactured by Schenectady International, Inc.

Example 1 :

Flakes (small particles) of an aromatic polyester copolymer 1 and 30wt.% of glass fiber, 0.4wt.% of montan wax acid sodium salt and 0.2wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Example 2:

Flakes (small particles) of an aromatic polyester copolymer 1 and 30wt.% of talc, 0.8wt.% of sodium stearate and 1.0wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Example 3:

Flakes (small particles) of an aromatic polyester copolymer 1 and 20wt.% of talc and 10wt.% of glass fiber, 0.8wt.% of sodium stearate and 1.0wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Example 4:

Flakes (small particles) of an aromatic polyester copolymer 2 and 30wt.% of glass fiber, 0.8wt.% of sodium stearate and 0.5 wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Example 5:

Flakes (small particles) of an aromatic polyester copolymer 3 and 30wt.% of glass fiber, 0.8wt.% of sodium stearate and 0.5 wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The

melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Comparative Example A

Flakes (small particles) of an aromatic polyester copolymer 1 and 30wt.% of glass fiber were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Comparative Example B:

Flakes (small particles) of an aromatic polyester copolymer 1 and 30wt.% of glass fiber, 0.4wt.% of montan wax acid sodium were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 2.

Comparative Example C:

Flakes (small particles) of an aromatic polyester copolymer 1 and 30wt.% of kaolin were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at die temp. 120°C. The physical properties are shown in Table 1.

Table 1 Physical properties of aromatic polyester moldings

	Example 1	Example 2	Example 3	Example 4	Example 5
Polymer	1)	1)	1)	2)	3)
additives	GF 30% Mon.Na 0.4% PTS 0.2%	talc 30% St.Na 0.8% PTS 1.0%	talc 20% GF 10% St.Na 0.8% PTS 1.0%	GF 30% St.Na 0.8% PTS 0.5%	GF 30% St.Na 0.8% PTS 0.5%
Moldability	good	good	good	good	good
tensile strength @ break (kgf/mm ²)	12.2	4.6	7.0	9.2	7.4
tensile elongation(%)	1.7	1.4	1.5	3.1	3.1
flexural strength (kgf/mm ²)	19.8	9.1	11.2	14.7	13.3
flexural modulus (kgf/mm ²)	1070	730	880	824	750
Izod impact strength (cm*kgf/cm)	7.1	2.2	3.7	7.8	6.3
HDT(die temp.120°C)	178	80	168	175	163

compost degradability	YES	YES	YES	YES	YES
T _{max} (min. at 120°C)	0.8	0.5	0.5	-	-

	Comp.ex.A	Comp.ex.B	Comp.ex.C
Polymer	1)	1)	1)
additives	GF 30%	GF 30% Mon.Na 0.4%	kaolin 30%
Moldability	poor	poor	poor
tensile strength @ break (kgf/mm ²)	10.3	10.8	7.8
tensile elongation(%)	3.8	3.3	3.0
flexural strength (kgf/mm ²)	18.5	18.6	9.4
flexural modulus (kgf/mm ²)	990	1000	560
Izod impact strength (cm*kgf/cm)	7.4	8.0	2.5
HDT	poor moldability	poor moldability	poor moldability
compost degradability	YES	YES	YES
Tmax (min. at 120°C)	1.8	1.3	1.3

It can be seen from the data in Table 1 that copolyester 1) in Example 1 to 3 achieves faster crystallization speed (by t max) than Comp.Ex.A-C by adding crystallization promotor and lubricant, which also provides good moldability and high HDT.

Copolyester 2) and 3) in Example 4 and 5 can also achieve high HDT by additions of both crystallization promotor and lubricant as seen in Table 1. In the following Examples, inorganic flame retardant or phosphorous-based flame retardant was blended with the compositions of the present invention. The pellets and test pieces were prepared as described in the same manner as that for Examples 1 - 5, above, and the results are reported in Table 2, below.

Examples 6:

Flakes (small particles) of an aromatic polyester copolymer 2 and 30wt.% of glass fiber, 18% of resorcinol bis(di-2,6-xylyl)phosphate(PX-200), 8% of phenolic polymer, 0.8wt.% of sodium stearate and 1.0 wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at a die temp. 120°C. The physical properties are shown in Table 2.

Example 7:

Flakes (small particles) of an aromatic polyester copolymer 2 and 30wt.% of $\text{Mg}(\text{OH})_2$, 30wt.% of glass fiber, 0.8wt.% of sodium stearate and 1.0 wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at a die temp. 120°C. The physical properties are shown in Table 2.

Comparative Example D:

Flakes (small particles) of an aromatic polyester copolymer 2 and 30wt.% of glass fiber, 20wt.% of talc, 0.8wt.% of sodium stearate and 1.0 wt.% of pentaerythrityl tetrastearate were compounded and pelletized. The pellet was then pre-dried and then melted at 200 mm diameter extruder at a cylinder temperature setting of 200 to 220°C. The melt was injection molded at a die temp. 120°C. The physical properties are shown in Table 2.

Table 2

	Example 6	Example 7	Comp. Ex. D
Polyester copolymer 2	42.2	38.2	48.2
PX-200	18.0		

Phenolic polymer	8.0		
Mg(OH) ₂		30	
Glass Fiber	30	30	30
Talc			20
Sodium Stearate	0.8	0.8	0.8
Pentaerythritol tetrastearate	1.0	1.0	1.0
Tensile strength (kgf/mm ²)	6.3	8.6	7.7
Elongation at break (%)	1.2	1.0	2.4
Flexural strength (kgf/mm ²)	9.2	12.3	11.2
Flexural modulus (kgf/mm ²)	770	1330	760
Notched Izod impact strength (cm*kgf/cm)	5.1	7.4	5.6
Flame resistance (3.2 mm)	V-0	V-0	Out
Heat deflection temperature (°C)	144	163	180

It can be seen from the data in Table 2 that copolyester 2 can achieve flame resistance V-0 at 3.2mm at Example6 and 7 compared with Comp.Ex.D.

【書類名】 外国語要約書

1. Abstract

The present invention relates to an injection molded product formed of aromatic polyester molding composition and a method of manufacture thereof. The molded product is biodegradable and has excellent mechanical properties. The molding composition comprises (A) 20 to 98.8 wt % at least one polyester selected from the group consisting of:

an aromatic polyester copolymer(a) having repeating units comprising an acid component and a glycol component, wherein the acid component is about 50 to 90 mol% terephthalic acid, about 0.2 to about 6 mol% sulfonic acid metal salt, and about 4 to 49.8 mol% aliphatic dicarboxylic acid; the glycol component is about 50 to 99.9 mol % ethylene glycol and about 0.1 to 50 mol% diethylene glycol;

an polyester copolymer (b) prepared by copolymerization with the first polyester with polyethylene glycol,

a branched polyester copolymer (c) by polycondensation of the first polyester copolymer with polyethylene glycol. And,

a polyester copolymer (d) having repeating units comprising aromatic dicarboxylic acids and glycol components; with proviso that the mole% of said aromatic dicarboxylic acids of said polyester copolymer is less than the mol % of the carboxylic acid content of said copolymer (a), (b), and (c),

(B) 1 to 60 wt% materials selected from the group consisting of reinforcing and filling materials,

(C) 0.1 to 7 wt% crystallization promotor, and,

(D) 0.1 to 5 wt% lubricant.

The injection molded product made from molding composition further comprising 1 to 60 wt% inorganic flame retardant, and methods for manufacturing an injection molded products using such molding compositions. The molded product has excellent heat distortion and relatively faster crystallization speed.

2. Representative Drawing

None

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